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PATENT  
0965-0232P

IN THE U.S. PATENT AND TRADEMARK OFFICE

In re application of

Before the Board of Appeals

Toshiro NISHI et al.

Appeal No.

Appl. No.: 09/118,833

Group: 1745

Filed: July 20, 1998

Examiner: J. Crepeau

For: SOLID ELECTROLYTE TYPE FUEL  
BATTERY

**APPEAL BRIEF**

Assistant Commissioner for Patents  
Washington, DC 20231

Sir:

In response to the Examiner's Office Action dated July 16, 2002, the following Appeal Brief is respectfully submitted in connection with the above-identified application.

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IN THE U.S. PATENT AND TRADEMARK OFFICE

In re application of	Before the Board of Appeals
Toshiro NISHI et al.	Appeal No.
Appl. No.: 09/118,833	Group: 1745
Filed: July 20, 1998	Examiner: J. Crepeau
Conf. No.: 9403	
For: SOLID ELECTROLYTE TYPE FUEL BATTERY	

BRIEF ON APPEAL ON BEHALF OF APPELLANTS FILED UNDER  
PROVISIONS OF 37 C.F.R. § 1.192

Honorable Commissioner  
of Patents  
Washington, D.C. 20231

Dear Sir:

This is an Appeal from the Final Rejection of July 16, 2002 of claims 4-28 in the above-identified application.

**I. REAL PARTY IN INTEREST**

As evidenced by the Assignment filed July 20, 1998, and recorded at Reel 9351, Frames 0059-0063 the Real Party In Interest in connection with the present application is the Assignee of record, MITSUBISHI HEAVY INDUSTRIES, LTD.

**II. RELATED APPEALS AND INTERFERENCES**

There are no pending Appeals or Interferences related to the present application known to Appellant or Appellant's Legal Representatives.

**III. STATUS OF CLAIMS**

Claims 4-28 are pending in the application. Claims 4-28 stand rejected.

**IV. STATUS OF AMENDMENTS**

An Amendment Under 37 C.F.R. § 1.111 was filed on February 28, 2000. An Amendment filed on July 10, 2000 was entered by the Advisory Action mailed July 21, 2000. An Amendment Under 37 C.F.R. § 1.116 was filed on September 11, 2000 and was refused entry in the Advisory Action mailed September 19, 2000. A Reply Under 37 C.F.R. § 1.116 was filed on February 12, 2001, that presented a Declaration Under 37 C.F.R. § 1.132 by Toshiro Nishi. A Continued Prosecution Application Under 37 C.F.R. § 1.53(d) was filed on May 10, 2001. A Preliminary Amendment was filed on August 7, 2001. An Amendment Under 37 C.F.R. § 1.111 (which presented appealed claims 4-28 in their final form as presented on Appeal) was filed on April 11, 2002. A Reply Under 37 C.F.R. § 1.116 was filed on November 18, 2002, that presented a second Declaration Under 37 C.F.R. § 1.132 by Toshiro Nishi, which was considered and entered by the Advisory Action mailed November 29, 2002. A Supplemental Reply Under 37

C.F.R. § 1.116 was filed on March 13, 2003, that presented a third Declaration Under 37 C.F.R. § 1.132 by Toshiro Nishi, which was considered and entered by the Advisory Action mailed March 27, 2003.

## **V. SUMMARY OF INVENTION**

The present invention as recited in claim 4 pertains to a solid electrolyte fuel battery, in which a sintered interconnector is used for connecting cells of the solid electrolyte fuel battery, and the sintered interconnector comprises a material having a matrix of the general formula  $MTiO_3$  where M is Mg, Ca, Sr, or Ba. The sintered interconnector is formed by, for instance, coating materials onto a surface and sintering them. (page 6, lines 15-22) The material of the interconnector can be  $MTiO_3$  where M is Mg, Ca, Sr or Ba. (page 5, lines 5-10)

In another embodiment of the invention as recited in claims 6 and 8, the present invention relates to a solid electrolyte fuel battery, in which a co-sintered interconnector for connecting cells of the solid electrolyte fuel battery comprises a material having a matrix of the general formula  $A_{1-x}B_xC_{1-y}D_yO_3$  where A is Ca, Sr or Ba (claim 6) or Mg (claim 8), B is a rare earth element, aluminum or chromium, C is titanium, D is niobium or tantalum,  $0 < x \leq 0.2$  and  $0 \leq y \leq 0.2$ . That is the co-sintered interconnector is  $A_{1-x}B_xC_{1-y}D_yO_3$  where A is Mg (e.g. claim 8), or Ca, Sr or Ba (claim 6), B is a rare earth element, aluminum or chromium, C is titanium, D is niobium or tantalum,  $0 < x \leq 0.2$

and  $0 \leq y \leq 0.2$ . (page 17, lines 6-7; page 22, lines 15-18; page 25, line 20; page 30, lines 6-7, page 35, line 4; page 36, lines 13-15; page 39, lines 25-26).

In the invention, the interconnector can be integrally burned within the battery. (claims 12-23; page 5, line 28 to page 6, line 8) The integrally burning can be performed at low temperatures such as 1,300 to 1,400 °C. (claims 21-23; page 6, line 13) The interconnector can be a hermetic interconnector having a relative density of greater to or equal to 94%. (claim 24; page 15, line 15) In the interconnector, the current passage can be in the vertical direction. (claims 5, 7, 9, 18-20; page 5, lines 13-15)

## **VI. ISSUES**

The first issue presented for review is whether Soma (USP 5,411,767) suggests all of the limitations set forth in claims 4-11 and 24-28 to properly support a rejection under 35 U.S.C. § 103. The second issue presented for review is whether JP 8-50913 (JP '913) in view of Soma suggests all of the limitations set forth in claims 4-28 to properly support a rejection under 35 U.S.C. § 103.

These issues are divided into Groups I-XXI, which are separately argued below.

**VII. GROUPING OF CLAIMS**

Appellants submit that claims 4-28 do not stand or fall together. Instead, Appellants respectfully wish to group claims 4-28 as follows:

- Group I: claim 4;
- Group II: claim 5;
- Group III: claim 24;
- Group IV: claim 6;
- Group V: claim 7;
- Group VI: claim 25;
- Group VII: claim 8;
- Group VIII claim 9;
- Group IX claim 26;
- Group X claims 10 and 11;
- Group XI claim 27;
- Group XII claims 12 and 13;
- Group XIII claim 18;
- Group XIV claim 21;
- Group XV claims 14 and 15;
- Group XVI claim 19;
- Group XVII claim 22;
- Group XVIII claims 16 and 17;



Group XIX claim 20;  
Group XX claim 23; and  
Group XXI claim 28.

## VIII. ARGUMENT

### A. Group I, Independent Claim 4

Soma fails to suggest all of the elements set forth in claim 4 to properly support a rejection under 35 U.S.C. § 103. JP '913 in view of Soma fails to suggest all of the elements set forth in claim 4 to properly support a rejection under 35 U.S.C. § 103. These failures of the cited art are accompanied by the Examiner's failure to acknowledge the clear advantages of the invention set forth in three declarations of unexpected results.

#### A.1. The Present Invention and its Advantages

The inventors have produced a solid electrolyte fuel battery, "in which a sintered interconnector is used for connecting cells of the solid electrolyte fuel battery, and the sintered interconnector comprises a material having a matrix of the general formula  $\text{MTiO}_3$  where M is Mg, Ca, Sr, or Ba." (claim 4) The inventive sintered  $\text{MTiO}_3$  matrix can be easily burned and has a small difference in expansion between oxidizing conditions and reducing conditions

when used as a fuel cell interconnector. The resulting solid electrolyte fuel battery has excellent durability and reliability, and the battery can be easily produced. See page 5, lines 16-23 of the specification.

Sintering provides important advantages to the invention. The claimed solid electrolyte fuel battery may be produced using ordinary manufacturing methods such as dipping, coating, slip casting or printing.

**A.2. Soma and JP '913**

**A.2.1 Soma**

Soma pertains to a method for producing an interconnector for a solid electrolyte type fuel cell. The Abstract of Soma states: "An interconnector material such as a perovskite complexed oxide is thermally sprayed onto the surface of an electrode of a solid electrolyte type fuel cell by plasma thermal spraying process at a temperature of not lower than 1,250 °C."

Soma fails to teach or suggest a "sintered interconnector" such as is set forth in claim 4.

At paragraph 2, lines 12-16 of the Final Office Action mailed July 16, 2002 (Paper No. 24), the Examiner admits to the failings of Soma, stating: "The reference does not expressly teach that the battery is co-sintered or that the interconnector is integrally burned within the battery, or that the current passage of the interconnector is current collection in the vertical direction

(claims 5, 7, 9). The reference further does not teach the same subscript ranges for the  $(La_{1-x}D_x)_{1-u}B_{1-w}_3$  compounds as recited in instant claims 6 or 8.” The Examiner then asserts that these limitations would be obvious to one having ordinary skill.

However, the Examiner has alleged obviousness over Soma not by combining references, but over a single reference. To establish a *prima facie* case of obviousness, “the prior art reference (or references when combined) must teach or suggest all the claim limitations.” MPEP §2142. In addition, if a reference needs to be modified to achieve the claimed invention “there must be a showing of a suggestion or motivation to modify the teachings of that reference to the claimed invention in order to support the obviousness conclusion.” Sibia Neurosciences Inc. v. Cadus Pharmaceutical Corp., 225 F. 3d 1349, 1356, 55 USPQ2d 1927, 1931 (Fed. Cir. 2000). As a result, the Examiner has failed to show how Soma teaches or suggests all the claim limitations.

Further, at paragraph 2, lines 29-31 of Paper No. 24, the Examiner states “Additionally, regarding the ‘co-sintered’ and ‘integrally burned’ limitations in the claims, these limitations are not considered to patentably distinguish over the Soma reference. These limitations are essentially process limitations, and therefore allow the claims to be interpreted as product-by-process claims.”

However, *Webster's Encyclopedic Unabridged Dictionary of the English Language* (Random House, 1996) provides relevant definitions of "sinter" as being "the product of a sintering operation" or "to bring about agglomeration (in metal particles) by heating." Also, an Applicant can be his own lexicographer. *See, e.g., In re Paulsen*, 30 F.3d 1475, 1480, 31 USPQ2d 1671, 1674 (Fed. Cir. 1994). Further, "during patent prosecution when claims can be amended, ambiguities should be recognized, scope and breath of language explored and clarification imposed." *In re Zletz*, 893 F.2d 319, 321, 13 USPQ2d 1320, 1323 (Fed. Cir. 1989).

Also, as known to routineers in the art, sintered materials can be readily discerned by fused particles observable in SEM (scanning electron microscope) photomicrographs that can be found for example on the Internet at [www.Osmonics.com/products/page764.htm](http://www.Osmonics.com/products/page764.htm) or [www.avxcorp.com/docs/technoinf/bscant.pdf](http://www.avxcorp.com/docs/technoinf/bscant.pdf).

As a result, the Applicants have clearly provided clarification of the language to amply demonstrate that the thermally sprayed material of Soma is fundamentally different from the sintered or co-sintered material of the invention.

Yet further, the Soma technology is based upon thermal spraying while the present invention uses sintering. Therefore, the principal of operation of Soma must be changed in order to use the reference to allege obviousness. If the

proposed modification of the prior art would change the principal of operation of the prior art invention being modified, then the teachings of the references are not sufficient to render the claims *prima facie* obvious. In re Ratti, 270 F.2d 810, 123 USPQ 349 (CCPA 1959). See also MPEP 2143.01.

As has been shown, the single reference of Soma fails to allege *prima facie* obviousness over claim 4 of Group I.

#### **A.2.2 JP'913 In View Of Soma**

JP '913 pertains to a solid electrolyte type fuel cell in which the air electrode and the interconnector can be simultaneously molded. JP '913 fails to disclose the material of the interconnector. The Examiner admits this failure in paragraph 3 of Paper No. 24.

In the conventional fuel battery shown in Figs. 5 and 6 of JP '913, a solid electrolyte layer 14 is formed at the outer circumference of the cylinder-shaped air electrode 13, and a fuel electrode is formed on the outer frame of the solid electrolyte layer 14. Fig. 1 of JP '913 shows an interconnector 24 molded to a part of the cylinder portion of the air electrode that concurrently acts as a support tube. Although this structure may achieve a small-size, lightweight fuel battery, the JP '913 reference merely discloses the feature of integrally sintering electrodes in the manufacturing process of the fuel battery.

The Examiner then turns to the teachings of Soma for the materials of the interconnector. However, the inability of Soma to be utilized to allege *prima facie* obviousness has been discussed above.

Also, to establish a *prima facie* case of obviousness, it is necessary for the Examiner to present evidence, preferably in the form of some teaching, suggestion, incentive, or inference in the applied prior art, or in the form of generally available knowledge, that one having ordinary skill in the art would have been lead to use the relevant teachings of the applied references in the proposed manner asserted by the Examiner to arrive at the invention. *See Ex parte Levengood*, 28 USPQ2d 1300 (BPAI 1993). Because the Examiner bears the initial burden of presenting a *prima facie* case of obviousness, if this burden is not met, then the burden of coming forth with evidence or argument does not shift to the Applicant. *In re Rijckaert*, 9 F.3d 1531, 28 USPQ2d 1955 (Fed. Cir. 1993). Likewise, where an Examiner fails to establish a proper *prima facie* case, the rejection is improper, and should be overturned. *In re Fine*, 837 F.2d 1071, 5 USPQ2d 1596 (Fed. Cir. 1988).

Further, the rigorous burden placed upon the Examiner for establishing *prima facie* obviousness has recently been emphasized by the United States Court of Appeals for the Federal Circuit in *In re Lee*, 277 F.3d 1338, 61 USPQ2d 1430 (Fed. Cir. 2002). In *Lee*, the court states:

As applied to the determination of patentability vel non when the issue is obviousness, "it is fundamental that rejections under 35 U.S.C. §103 must be based on evidence comprehended by the language of that section." In re Grasselli, 713 F.2d 731, 739, 218 USPQ 769, 775 (Fed. Cir. 1983). The essential factual evidence on the issue of obviousness is set forth in Graham v. John Deere Co., 383 U.S. 1, 17-18, 86 S. Ct. 684, 15 L.Ed.2d 545, 148 USPQ 459, 467 (1966) and extensive ensuing precedent. The patent examination process centers on prior art and the analysis thereof. When patentability turns on the question of obviousness, the search for and analysis of the prior art includes evidence relevant to the finding of whether there is a teaching, motivation, or suggestion to select and combine the references relied on as evidence of obviousness. See, e.g., McGinley v. Franklin Sports, Inc., 262 F.3d 1339, 1351-52, 60 USPQ2d 1001, 1008 (Fed. Cir. 2001) ("the central question is whether there is reason to combine [the] references," a question of fact drawing on the Graham factors).

"The factual inquiry whether to combine references must be thorough and searching." Id. It must be based on objective evidence of record. This precedent has been reinforced in myriad decisions, and cannot be dispensed with. See, e.g., Brown & Williamson Tobacco Corp. v. Philip Morris Inc., 229 F.3d 1120, 1124-25, 56 USPQ2d 1456, 1459 (Fed. Cir. 2000) ("a showing of a suggestion, teaching, or motivation to combine the prior art references is an 'essential component of an obviousness holding'") (quoting C.R. Bard, Inc. v. M3 Systems, Inc., 157 F.3d 1340, 1352, 48 USPQ2d 1225, 1232 (Fed. Cir. 1998)); In re Dembiczak, 175 F.3d 994, 999, 50 USPQ2d 1614, 1617 (Fed. Cir. 1999) ("Our case law makes clear that the best defense against the subtle but powerful attraction of a hindsight-based obviousness analysis is rigorous application of the requirement for a showing of the teaching or motivation to combine prior art references."); In re Dance, 160 F.3d 1339, 1343, 48 USPQ2d 1635, 1637 (Fed. Cir. 1998) (there must be some motivation, suggestion, or teaching of the desirability of making the specific combination that was made by the applicant); In re Fine, 837 F.2d 1071, 1075, 5 USPQ2d 1596, 1600 (Fed. Cir. 1988) ("teachings of references can be combined only if there is some suggestion or incentive to do so.") (emphasis in original) (quoting ACS Hosp. Sys., Inc. v. Montefiore Hosp., 732 F.2d 1572, 1577, 221 USPQ 929, 933 (Fed. Cir. 1984)).

The need for specificity pervades this authority. See, e.g., In re Kotzab, 217 F.3d 1365, 1371, 55 USPQ2d 1313, 1317 (Fed. Cir. 2000) ("particular findings must be made as to the reason the skilled artisan, with no knowledge of the claimed invention, would have selected these components for combination in the manner claimed"); In re Rouffet, 149 F.3d 1350, 1359, 47 USPQ2d 1453, 1459 (Fed. Cir. 1998) ("even when the level of skill in the art is high, the Board must identify specifically the principle, known to one of ordinary skill, that suggests the claimed combination. In other words, the Board must explain the reasons one of ordinary skill in the art would have been motivated to select the references and to combine them to render the claimed invention obvious."); In re Fritch, 972 F.2d 1260, 1265, 23 USPQ2d 1780, 1783 (Fed. Cir. 1992) (the examiner can satisfy the burden of showing obviousness of the combination "only by showing some objective teaching in the prior art or that knowledge generally available to one of ordinary skill in the art would lead that individual to combine the relevant teachings of the references").

In re Lee, 277 F.3d at 1342-1343, 61 USPQ2d at 1433-1434 (emendation in the original).

The Examiner uses the teachings of Soma for material components. However, the Examiner fails to realize that Soma represents a fundamentally different technology. A prior art reference is analogous if the reference is in the field of Appellants' endeavor or, if not, the reference is reasonably pertinent to the particular problem with which the inventor was concerned. "In order to rely on a reference as a basis for rejection of the applicant's invention, the reference must either be in the field of the applicant's endeavor or, if not, then be reasonably pertinent to the particular problem with the which the inventor was concerned." In re Oetiker, 977 F.2d 1443, 1447, 24 USPQ2d 1443, 1445 (Fed.



Cir. 1992). However, the field of endeavor of Soma is thermal spraying, which is in marked contrast to the sintering of the present invention. Thus, Soma is non-analogous art. As a result, the Examiner has relied upon impermissible hindsight reconstruction, discussed above, to combine Soma with JP '913.

Thus, a case of *prima facie* obviousness has not been made over JP '913 in view of Soma.

#### **A.2.3 Unexpected Results**

Even if it is assumed arguendo, that *prima facie* obviousness can be alleged over Soma or the combination of JP '913 with Soma, this obviousness would be fully rebutted by the three showings of unexpected results submitted as Declarations Under 37 C.F.R. §1.132.

The first Declaration was filed on February 12, 2002. The first Declaration clearly describes the inventive process of sintering compared to the Soma-type thermal spraying process. The appendices to this first Declaration included overheads describing the sintering process and two publications pertaining to plasma spraying. The Declaration use  $\text{LaCrO}_3$  as the exemplary material. Comparative result between sintering and thermal spraying were summarized in a table, which is reproduced as Table 1, below.

Table 1. Comparison of thermal spraying and sintering in fuel cell preparation.

	Fuel Cell Prepared by Thermal Spraying Process	Fuel Cell Prepared by Sintering Process (Present Invention)
Required time for production	150 min/fuel cell	15 min/fuel cell
Yield on materials	3 - 10%	90% or more
Equipment cost	Basic Amount	1/10
Construction Cost	Basic Amount	1/5
Materials Cost	Basic Amount	1/2
Cell production cost	1/3 million yen/kw	50,000 yen/kw

Despite the clear advantages of the invention, the Examiner did not allow the application. In the Advisory Action mailed February 28, 2001 (Paper No. 16), the Examiner stated:

Independent claims 6 and 8 do not recite the process limitations that allegedly produce the unexpected results of the invention, therefore the declaration and arguments relating to these limitations are not commensurate in scope with these claims. Results in Table on page 4 of the declaration are only shown for a prior art composition of lanthanum chromite. However, lanthanum chromite is not germane to the outstanding rejection. A comparison between the process of Soma and the process of the present invention should be made using the materials relied upon in Soma.

That is, the Examiner failed to realize that clear advantages are realized for the sintering of any suitable material compared to the Soma-type thermal spraying.

On November 18, 2002, a second Declaration was placed before the Examiner. This second Declaration submitted SEM photomicrographs of a

plasma sprayed material (A) and a co-sintered material (B). The Board of Appeals is respectfully requested to refer to the original blue photomicrographs in the originally filed declaration(s) in order to observe the dramatic differences in materials produced by the two different processes. The declaration at paragraph 3 states:

When plasma spraying is applied as in SOMA and other conventional art processes, a number of air spaces are produced in the plasma-sprayed film having a thickness of from several to dozens of microns. This can be clearly observed in the dense-blue cored portion of SEM photomicrograph (A) (also note the micron bar for scale).

In contrast, when the inventive co-sintering is utilized, a film having a very dense structure is formed. This is clearly observable in SEM photomicrograph (B). Incidentally, the portions that appear to be air spaces in SEM photomicrograph (B) are produced during thermal etching. However, the sizes of these air spaces are very small, on the order of submicrons, as indicated by the micron bar for scale.

As a result, a "sintered" or "co-sintered" material such as an interconnector does not represent a process step. Instead, a "sintered" or "co-sintered" material represents a physical state of matter that is achieved through the sintering process. This state of matter is readily discernable by comparing SEM photomicrographs (A) and (B).

Therefore, the plasma sprayed film of SOMA is incapable of attaining the sintered or co-sintered interconnector of the invention. Therefore, SOMA fails to either teach or suggest a sintered or co-sintered interconnector of the invention. The high density achievable by the inventive technology and demonstrated in the attached SEM photomicrographs are a clear demonstration of unexpected results over SOMA.

Despite the clear differences between the invention and Soma in the second Declaration, the Examiner was not convinced. In the Advisory Action mailed November 29, 2002 (Paper No. 27), the Examiner states that "the declaration does not appear to state that the materials being compared have the same chemical composition. This would be required for a meaningful comparison."

In the Advisory Action, the Examiner then stated something curious: "Soma discloses a 'heat treatment' step of at least 1250 degrees C after the step of plasma spraying (col. 2, lines 47-57 and col. 6, lines 44-50 of Soma). The declaration does not appear to account for this additional heat treatment step. This step is believed to be critical to the comparison of the process of Soma and the claimed process, as it is essentially a 'sintering' step . . ." However, if this heat treatment step is a sintering step, why use thermal spraying at all? That is, the Examiner is inferring that two discrete steps are needed in Soma to form the material. In contrast, the present invention only requires one step: sintering: Therefore, the Examiner is either 1) verifying that Soma uses a fundamentally different process from the invention, or 2) the principal of operation must be changed. See *In re Ratti, supra*. In either case, the rejections must fall by the Examiner's own admissions.

However, to further expedite prosecution, a third Declaration was filed on March 13, 2003. This Declaration resubmitted SEM photomicrographs and in

paragraph 3 clearly stated "The materials (A) and (B) being compared have the same chemical composition." Therefore, the comparison between Soma and the invention is clear.

On March 27, 2003, the Examiner mailed an Advisory Action (paper No. 30). In the Advisory Action, the Examiner still tried to equate the heat treatment step of Soma with the sintering of the invention. The Examiner then asserted that even if the Declarations would overcome Soma as a primary reference, Soma could still be used as a secondary reference. However, the inability of Soma to be used to allege *prima facie* obviousness, e.g., by being non-analogous art, has been discussed in detail above. Further, the unexpected results in the three declarations are directed at the Soma reference.

Finally, it should be noted that the material recited in the claims have already been employed in a solid oxide fuel cell and has been shown to provide outstanding results as a new material. The materials of the present invention have been evaluated at International and Japanese Science Conferences by others, for example, the 9th Cimtec World Forum on New Materials (1999).

As a result, applicants believe that the evidence has overcome Soma alone or in any combination.

**A.3 Summary**

As has been shown, the Examiner has failed to establish a *prima facie* case of obviousness over Soma or the combination of Soma and JP '913. Further, three declarations have clearly demonstrated unexpected results over the applied prior art. Appellants therefore respectfully submit that the combination of limitations as set forth in independent claim 4 of Group I is not obvious by Soma or the combination of Soma and JP '913, for the reasons explained above.

Accordingly, reversal of the Examiner's rejection based on the above arguments is respectfully requested.

**B. Group II, Claim 5**

Soma or the combination of Soma and JP '913 fails to suggest all of the limitations set forth in claim 5 to properly support the rejections of Group II under 35 U.S.C. § 103.

Soma and JP '913 have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition. Also, claim 5 depends upon claim 4 (Group I), and all of the distinctions of the invention over Group I are equally applicable to Group II.

Claim 5 further recites the current passage of the interconnector is current collection in the vertical direction from a fuel electrode through the

interconnector. This embodiment is supported at page 5, lines 13-15 of the specification.

In paragraph 5 of the Office Action mailed April 10, 2000 (Paper No. 8), the Examiner asserts that "the direction of current collection is a design choice that may be manipulated according to the needs of the artisan." The Examiner repeats this assertion in paragraph 4 of the Office Action mailed October 26, 2001 (paper No. 21).

However, as noted at page 8 of the Amendment filed July 10, 2002, vertical collection of current would not be an obvious design choice for one of ordinary skill in the art of producing fuel cells. Figures 44(a) and 44(b) of the instant invention depict vertical and horizontal current collection. As shown in Figure 44(a) and described on page 34, lines 8-10 of the specification, vertical current collection has advantages because high resistance can be evaded by the thinness of the interconnector. In vertical collection of current, the air electrode 15 is located just above the fuel electrode 12 via the interconnector 14. By thinning the layer of the interconnector, overvoltage due to resistance of the interconnector is decreased.

In contrast, Figure 44(b) of the application shows that current collection in the horizontal direction is disadvantageous because it is not applicable for high resistance material. In horizontal current collection, current needs to be passed through the thin film in the same direction as the axial direction. Thus, a

decrease in overvoltage cannot be achieved.

As discussed above, horizontal current collection and vertical current collection are different processes which possess different properties. Thus, the vertical direction of the fuel cells of the instant invention is not an arbitrary design choice.

Therefore, claim 5 of Group II is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejections based on the claimed combination of limitations and the above arguments is respectfully requested.

**C. Group III, Claim 24**

Soma or the combination of Soma and JP '913 fails to suggest all of the elements set forth in claim 24 to properly support the rejections of Group II under 35 U.S.C. § 103.

Soma and JP '913 have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition. Also, claim 24 depends upon claim 4 (Group I), and all of the distinctions of the invention over Group I are equally applicable to Group III.

Claim 24 contains the additional distinction that the interconnector is a hermetic interconnector having a relative density of greater of equal to 94%. In paragraph 3 of the Office Action mailed July 17, 2002 (Paper No. 24), the Examiner asserts "Soma et al. teach interconnectors having relative densities of



at least 95% and which comprise perovskite materials that are not patentably distinct from the instantly claimed materials.” Soma at column 9, lines 61-63 states: “It is possible to obtain a relative density of more than 95% according to the invention.”

However, Soma pertains to thermal spraying. Soma contains no teaching or suggestion pertaining to how high densities can be obtained in a sintered material.

Therefore, claim 24 of Group III is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejections based on the claimed combination of limitations and the above arguments is respectfully requested.

*D. Group IV, Claim 6*

Soma fails to suggest all of the elements set forth in claim 6 to properly support a rejection under 35 U.S.C. § 103. JP '913 in view of Soma fails to suggest all of the elements set forth in claim 6 to properly support a rejection under 35 U.S.C. § 103. These failures of the cited art are accompanied by the Examiner's failure to acknowledge the clear advantages of the invention set forth in three declarations of unexpected results.

Soma and JP '913 have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition.

Claim 6 is similar to claim 4 of Group I, except that the material of the interconnector "comprises a material having a matrix of the general formula  $A_{1-x}B_xC_{1-y}D_yO_3$  where A is Ca, Sr or Ba, B is a rare earth element, aluminum or chromium, C is titanium, D is niobium or tantalum,  $0 < x \leq 0.2$  and  $0 \leq y \leq 0.2$ ."

In paragraph 4 of the Office Action mailed October 26, 2001, the Examiner admits the failure of the prior art of Soma to disclose the formula of claim 6 (and claim 8). The Examiner then asserts "Regarding the subscript ranges of the  $(La_{1-x}D_x)_{1-u}B_{1-w}O_3$  material, these ranges have not been shown to be critical variables in the practice of the invention. Applicant must show that the particular subscript ranges are critical, generally by showing that the claimed ranges achieve unexpected results relative to the prior art ranges." (citation omitted).

However, Fig. 30 of the application shows the criticality of the  $x = 0.2$  limitation, and this result is directly opposite the teachings at column 4, lines 44 and 45 of Soma, which shows no criticality for an 0.2 fraction. A prior art reference must be considered in its entirety, i.e., as a whole, including portions that would lead away from the claimed invention. W.L. Gore & Associates, Inc. v. Garlock, Inc., 721 F.2d 1540, 220 USPQ 303 (Fed. Cir. 1983).

Therefore, claim 6 of Group IV is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejections based on the claimed

combination of limitations and the above arguments is respectfully requested.

*E. Group V, Claim 7*

Soma or the combination of Soma and JP '913 fails to suggest all of the elements set forth in claim 7 to properly support the rejections of Group V under 35 U.S.C. § 103.

Soma and JP '913 have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition. Also, claim 7 depends upon claim 6 (Group IV), and all of the distinctions of the invention over Group IV are equally applicable to Group V.

Claim 7 further recites the current passage of the interconnector is current collection in the vertical direction from a fuel electrode through the interconnector. This embodiment is supported at page 5, lines 13-15 of the specification.

In paragraph 5 of the Office Action mailed April 10, 2000 (Paper No. 8), the Examiner asserts that "the direction of current collection is a design choice that may be manipulated according to the needs of the artisan." The Examiner repeats this assertion in paragraph 4 of the Office Action mailed October 26, 2001 (paper No. 21).

However, as noted at page 8 of the Amendment filed July 10, 2002, vertical collection of current would not be an obvious design choice for one of

ordinary skill in the art of producing fuel cells. Figures 44(a) and 44(b) of the instant invention depict vertical and horizontal current collection. As shown in Figure 44 (a) and described on page 34, lines 8-10 of the specification, vertical current collection has advantages because high resistance can be evaded by the thinness of the interconnector. In vertical collection of current, the air electrode 15 is located just above the fuel electrode 12 via the interconnector 14. By thinning the layer of the interconnector, overvoltage due to resistance of the interconnector is decreased.

In contrast, Figure 44(b) of the application shows that current collection in the horizontal direction is disadvantageous because it is not applicable for high resistance material. In horizontal current collection, current needs to be passed through the thin film in the same direction as the axial direction. Thus, a decrease in overvoltage cannot be achieved.

As discussed above, horizontal current collection and vertical current collection are different processes which possess different properties. Thus, the vertical direction of the fuel cells of the instant invention is not an arbitrary design choice.

Therefore, claim 7 of Group V is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejections based on the claimed combination of limitations and the above arguments is respectfully requested.

**F. Group VI, Claim 25**

Soma or the combination of Soma and JP '913 fails to suggest all of the elements set forth in claim 25 to properly support the rejections of Group VI under 35 U.S.C. § 103.

Soma and JP '913 have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition. Also, claim 25 depends upon claim 6 (Group IV), and all of the distinctions of the invention over Group IV are equally applicable to Group VI.

Claim 25 contains the additional distinction that the interconnector is a hermetic interconnector having a relative density of greater or equal to 94%. In paragraph 3 of the Office Action mailed July 17, 2002 (Paper No. 24), the Examiner asserts "Soma et al. teach interconnectors having relative densities of at least 95% and which comprise perovskite materials that are not patentably distinct from the instantly claimed materials." Soma at column 9, lines 61-63 states: "It is possible to obtain a relative density of more than 95% according to the invention."

However, Soma pertains to thermal spraying. Soma contains no teaching or suggestion pertaining to how high densities can be obtained in a sintered material.

Therefore, claim 25 of Group VI is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejections based on the claimed

combination of limitations and the above arguments is respectfully requested.

G. Group VII, Claim 8

Soma fails to suggest all of the elements set forth in claim 8 to properly support a rejection under 35 U.S.C. § 103. JP '913 in view of Soma fails to suggest all of the elements set forth in claim 8 to properly support a rejection under 35 U.S.C. § 103. These failures of the cited art are accompanied by the Examiner's failure to acknowledge the clear advantages of the invention set forth in three declarations of unexpected results.

Soma and JP '913 have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition.

Claim 8 is similar to claim 4 of Group I and claim 6 of Group IV, except that the material of the interconnector "comprises a material having a matrix of the general formula  $A_{1-x}B_xC_{1-y}D_yO_3$  where A is Mg, B is a rare earth element, aluminum or chromium, C is titanium, D is niobium or tantalum,  $0 < x \leq 0.2$  and  $0 \leq y \leq 0.2$ ."

In paragraph 4 of the Office Action mailed October 26, 2001, the Examiner admits the failure of the prior art of Soma to disclose the formula of claim 8 (and claim 6). The Examiner then asserts "Regarding the subscript ranges of the  $(La_{1-x}D_x)_{1-u}B_{1-w}O_3$  material, these ranges have not been shown to

be critical variables in the practice of the invention. Applicant must show that the particular subscript ranges are critical, generally by showing that the claimed ranges achieve unexpected results relative to the prior art ranges.” (citation omitted).

However, Fig. 30 of the application shows the criticality of the  $x = 0.2$  limitation, and this result is directly opposite the teachings at column 4, lines 44 and 45 of Soma. A prior art reference must be considered in its entirety, i.e., as a whole, including portions that would lead away from the claimed invention. W.L. Gore & Associates, Inc. v. Garlock, Inc., 721 F.2d 1540, 220 USPQ 303 (Fed. Cir. 1983), *cert denied*, 469 U.S. 851 (1984).

Therefore, claim 8 of Group VII is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejections based on the claimed combination of limitations and the above arguments is respectfully requested.

H. Group VIII, Claim 9

Soma or the combination of Soma and JP '913 fails to suggest all of the elements set forth in claim 9 to properly support the rejections of Group VIII under 35 U.S.C. § 103.

Soma and JP '913 have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition. Also, claim 9 depends upon claim 8 (Group VII), and all of the

distinctions of the invention over Group VII are equally applicable to Group VIII.

Claim 9 further recites the current passage of the interconnector is current collection in the vertical direction. This embodiment is supported at page 5, lines 13-15 of the specification.

In paragraph 5 of the Office Action mailed April 10, 2000 (Paper No. 8), the Examiner asserts that "the direction of current collection is a design choice that may be manipulated according to the needs of the artisan." The Examiner repeats this assertion in paragraph 4 of the Office Action mailed October 26, 2001 (paper No. 21).

However, as noted at page 8 of the Amendment filed July 10, 2002, vertical collection of current would not be an obvious design choice for one of ordinary skill in the art of producing fuel cells. Figures 44(a) and 44(b) of the instant invention depict vertical and horizontal current collection. As shown in Figure 44(a) and described on page 34, lines 8-10 of the specification, vertical current collection has advantages because high resistance can be evaded by thinness of the interconnector. In vertical collection of current, the air electrode 15 is located just above the fuel electrode 12 via the interconnector 14. By thinning the layer of the interconnector, overvoltage due to resistance of the interconnector is decreased.

In contrast, Figure 44(b) of the application shows that current collection in the horizontal direction is disadvantageous because it is not applicable for high



resistance material. In horizontal current collection, current needs to be passed through the thin film in the same direction as the axial direction. Thus, a decrease in overvoltage cannot be achieved.

As discussed above, horizontal current collection and vertical current collection are different processes which possess different properties. Thus, the vertical direction of the fuel cells of the instant invention is not an arbitrary design choice.

Therefore, claim 9 of Group VIII is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejections based on the claimed combination of limitations and the above arguments is respectfully requested.

**I. Group IX, Claim 26**

Soma or the combination of Soma and JP '913 fails to suggest all of the elements set forth in claim 26 to properly support the rejections of Group IX under 35 U.S.C. § 103.

Soma and JP '913 have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition. Also, claim 26 depends upon claim 8 (Group VII), and all of the distinctions of the invention over Group VII are equally applicable to Group IX.

Claim 26 contains the additional distinction that the interconnector is a hermetic interconnector having a relative density of greater or equal to 94%. In

paragraph 3 of the Office Action mailed July 17, 2002 (Paper No. 24), the Examiner asserts: "Soma et al. teach interconnectors having relative densities of at least 95% and which comprise perovskite materials that are not patentably distinct from the instantly claimed materials." Soma at column 9, lines 61-63 states: "It is possible to obtain a relative density of more than 95% according to the invention."

However, Soma pertains to thermal spraying. Soma contains no teaching or suggestion pertaining to how high densities can be obtained in a sintered material.

Therefore, claim 26 of Group IX is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejections based on the claimed combination of limitations and the above arguments is respectfully requested.

**J. Group X, Claims 10-11**

Soma fails to suggest all of the elements set forth in claims 10-11 to properly support a rejection under 35 U.S.C. § 103. JP '913 in view of Soma fails to suggest all of the elements set forth in claims 10-11 to properly support a rejection under 35 U.S.C. § 103. These failures of the cited art are accompanied by the Examiner's failure to acknowledge the clear advantages of the invention set forth in three declarations of unexpected results.

Soma and JP '913 have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition.

The inventors have produced a solid electrolyte fuel battery, in which "an interconnector for connecting cells of the solid electrolyte fuel battery comprises a material having a matrix of the general formula  $MTiO_3$  where M is Mg, Ca, Sr, or Ba, wherein the interconnector is integrally burned within said battery. (claim 10)

Regarding the "integrally burned" embodiment, JP '913 pertains to a solid electrolyte fuel cell that is "integrally sintered." See English Abstract of JP '913. The Examiner admits that JP '913 fails to teach the material(s) that comprise the interconnector, or the temperature at which the sintering is performed. See, e.g., paragraph 3 of the Office Action mailed July 16, 2002 (Paper No. 24). The Examiner, then turns to Soma for teachings pertaining to materials.

However, the inability of Soma or JP '913 and Soma to suggest the present invention has been discussed in detail above, along with the showings of unexpected results. For brevity, the distinctions of the invention over Soma or JP '913 and Soma are not repeated here.

Therefore, claims 10-11 of Group X are patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejections based on the

claimed combination of limitations and the above arguments is respectfully requested.

K. Group XI, Claim 27

Soma or the combination of Soma and JP '913 fails to suggest all of the elements set forth in claim 27 to properly support the rejections of Group XI under 35 U.S.C. § 103.

Soma and JP '913 have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition. Also, claim 27 depends upon claim 10 (Group X), and all of the distinctions of the invention over Group X are equally applicable to Group XI

Claim 27 contains the additional distinction that the interconnector is a hermetic interconnector having a relative density of greater or equal to 94%. In paragraph 3 of the Office Action mailed July 17, 2002 (Paper No. 24), the Examiner asserts: "Soma et al. teach interconnectors having relative densities of at least 95% and which comprise perovskite materials that are not patentably distinct from the instantly claimed materials." Soma at column 9, lines 61-63 states: "It is possible to obtain a relative density of more than 95% according to the invention."

However, Soma pertains to thermal spraying. Soma contains no teaching or suggestion pertaining to how high densities can be obtained in a sintered

material.

Therefore, claim 27 of Group XI is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejections based on the claimed combination of limitations and the above arguments is respectfully requested.

L. Group XII, Claims 12 and 13

JP '913 in view of Soma fails to suggest all of the elements set forth in claims 10-11 to properly support a rejection under 35 U.S.C. § 103. These failures of the cited art are accompanied by the Examiner's failure to acknowledge the clear advantages of the invention set forth in three declarations of unexpected results.

Claims 12-13 are method claims that are clear of the obviousness rejection over Soma alone. Soma and JP '913 have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition.

Claim 12 is a method claim comprising integrally burning within the battery the interconnector for connecting cells of the solid electrolyte fuel battery. The battery and interconnector contain all the structural limitations of claim 4 Group I. As a result, all of the distinctions of the invention over Soma and JP '913 set forth for Group I are equally valid for claims 12 and 13 of Group XII. That is, JP '913 and Soma fail to support an allegation of *prima facie* obviousness

over claims 12-13. Even if obviousness can be alleged, unexpected results offer full rebuttal.

Accordingly, reversal of the Examiner's rejection based on the claimed combination of limitations and the above arguments is respectfully requested.

*M. Group XIII, Claim 18*

JP '913 and Soma fail to suggest all of the elements set forth in claim 18 to properly support the rejections of Group XIII under 35 U.S.C. § 103.

JP '913 and Soma have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition. Also, claim 18 depends upon claim 12 (Group XII), and all of the distinctions of the invention over Group XII are equally applicable to Group XIII.

Claim 18 further recites the current passage of the interconnector is current collection in the vertical direction. This embodiment is supported at page 5, lines 13-15 of the specification.

In paragraph 5 of the Office Action mailed April 10, 2000 (Paper No. 8), the Examiner asserts that "the direction of current collection is a design choice that may be manipulated according to the needs of the artisan." The Examiner repeats this assertion in paragraph 4 of the Office Action mailed October 26, 2001 (paper No. 21).

However, as noted at page 8 of the Amendment filed July 10, 2002, vertical collection of current would not be an obvious design choice for one of ordinary skill in the art of producing fuel cells. Figures 44(a) and 44(b) of the instant invention depict vertical and horizontal current collection. As shown in Figure 44(a) and described on page 34, lines 8-10 of the specification, vertical current collection has advantages because high resistance can be evaded by thinness of the interconnector. In vertical collection of current, the air electrode 15 is located just above the fuel electrode 12 via the interconnector 14. By thinning the layer of the interconnector, overvoltage due to resistance of the interconnector is decreased.

In contrast, Figure 44(b) of the application shows that current collection in the horizontal direction is disadvantageous because it is not applicable for high resistance material. In horizontal current collection, current needs to be passed through the thin film in the same direction as the axial direction. Thus, a decrease in overvoltage cannot be achieved.

As discussed above, horizontal current collection and vertical current collection are different processes which possess different properties. Thus, the vertical direction of the fuel cells of the instant invention is not an arbitrary design choice.

Therefore, claim 18 of Group XIII is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejection based on the claimed

combination of limitations and the above arguments is respectfully requested.

*N. Group XIV, Claim 21*

JP '913 and Soma fail to suggest all of the elements set forth in claim 21 to properly support the rejection of Group XIV under 35 U.S.C. § 103.

JP '913 and Soma have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition. Also, claim 21 depends upon claim 12 (Group XII), and all of the distinctions of the invention over Group XII are equally applicable to Group XIV.

Claim 21 further covers the embodiment that integrally burning is performed at a temperature of 1,300 °C to 1,400 °C. This advantageous temperature is discussed at page 6, lines 11-13 of the specification, which states: "This material can be burned not at a conventional high temperature of 1,600 °C, but at a lower temperature of 1,300 to 1,400 °C . . ."

In paragraph 3 of the Office Action mailed July 17, 2002 (Paper No. 24), the Examiner analogizes the heat treatment temperature of Soma to the inventive integrally burning step. However, the logical failures of this analogy have been discussed above. That is, if this heat treatment step of Soma is an integrally burning or sintering step, why use thermal spraying at all? That is, the Examiner is inferring that two discrete steps are needed in Soma to form the material. In contrast, the present invention only requires one step:



sintering. Therefore, the Examiner is either 1) verifying that Soma uses a fundamentally different process from the invention, or 2) the principal of operation must be changed. See In re Ratti. In either case, the rejection must fall by the Examiner's own admissions.

Therefore, claim 21 of Group XIV is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejection based on the claimed combination of limitations and the above arguments is respectfully requested.

O. Group XV, Claims 14 and 15

JP '913 in view of Soma fails to suggest all of the elements set forth in claims 14-15 to properly support a rejection under 35 U.S.C. § 103. These failures of the cited art are accompanied by the Examiner's failure to acknowledge the clear advantages of the invention set forth in three declarations of unexpected results.

Claims 14-15 are method claims that are clear of the obviousness rejection over Soma alone. Soma and JP '913 have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition.

Claim 14 is a method claim comprising integrally burning within the battery the interconnector for connecting cells of the solid electrolyte fuel battery. The battery and interconnector contain all the structural limitations of claim 6

of Group IV. As a result, all of the distinctions of the invention over Soma and JP '913 set forth for Group VI are equally valid for claims 14 and 15 of Group XV. That is, JP '913 and Soma fail to support an allegation of *prima facie* obviousness over claims 14-15. Even if obviousness can be alleged, unexpected results offer full rebuttal.

Accordingly, reversal of the Examiner's rejection based on the claimed combination of limitations and the above arguments is respectfully requested.

P. Group XVI, Claim 19

JP '913 and Soma fail to suggest all of the elements set forth in claim 19 to properly support the rejection of Group XVI under 35 U.S.C. § 103.

JP '913 and Soma have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition. Also, claim 19 depends upon claim 14 (Group XV), and all of the distinctions of the invention over Group XV are equally applicable to Group XVI.

Claim 19 further recites the current passage of the interconnector is current collection in the vertical direction. This embodiment is supported at page 5, lines 13-15 of the specification.

In paragraph 5 of the Office Action mailed April 10, 2000 (Paper No. 8), the Examiner asserts that "the direction of current collection is a design choice that may be manipulated according to the needs of the artisan." The Examiner

repeats this assertion in paragraph 4 of the Office Action mailed October 26, 2001 (paper No. 21).

However, as noted at page 8 of the Amendment filed July 10, 2002, vertical collection of current would not be an obvious design choice for one of ordinary skill in the art of producing fuel cells. Figures 44(a) and 44(b) of the instant invention depict vertical and horizontal current collection. As shown in Figure 44(a) and described on page 34, lines 8-10 of the specification, vertical current collection has advantages because high resistance can be evaded by the thinness of the interconnector. In vertical collection of current, the air electrode 15 is located just above the fuel electrode 12 via the interconnector 14. By thinning the layer of the interconnector, overvoltage due to resistance of the interconnector is decreased.

In contrast, Figure 44(b) of the application shows that current collection in the horizontal direction is disadvantageous because it is not applicable for high resistance material. In horizontal current collection, current needs to be passed through the thin film in the same direction as the axial direction. Thus, a decrease in overvoltage cannot be achieved.

As discussed above, horizontal current collection and vertical current collection are different processes which possess different properties. Thus, the vertical direction of the fuel cells of the instant invention is not an arbitrary design choice.

Therefore, claim 19 of Group XVI is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejection based on the claimed combination of limitations and the above arguments is respectfully requested.

Q. *Group XVII, Claim 22*

JP '913 and Soma fail to suggest all of the elements set forth in claim 22 to properly support the rejection of Group XIV under 35 U.S.C. § 103.

JP '913 and Soma have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition. Also, claim 22 depends upon claim 14 (Group XV), and all of the distinctions of the invention over Group XV are equally applicable to Group XVII.

Claim 22 further covers the embodiment that integrally burning is performed at a temperature of 1,300 °C to 1,400 °C. This advantageous temperature is discussed at page 6, lines 11-13 of the specification, which states: "This material can be burned not at a conventional high temperature of 1,600 °C, but at a lower temperature of 1,300 to 1,400 °C . . ."

In paragraph 3 of the Office Action mailed July 17, 2002 (Paper No. 24), the Examiner analogizes the heat treatment temperature of Soma to the inventive integrally burning step. However, the cognitive failures of this analogy have been discussed above. That is, if this heat treatment step of Soma is an integrally burning or sintering step, why use thermal spraying at



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all? That is, the Examiner is inferring that two discrete steps are needed in Soma to form the material. In contrast, the present invention only requires one step: sintering. Therefore, the Examiner is either 1) verifying that Soma uses a fundamentally different process from the invention, or 2) the principal of operation must be changed. *See In re Ratti, supra*. In either case, the rejection must fall by the Examiner's own admissions.

Therefore, claim 22 of Group XVII is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejection based on the claimed combination of limitations and the above arguments is respectfully requested.

*R. Group XVIII, Claims 16 and 17*

JP '913 in view of Soma fails to suggest all of the elements set forth in method claims 16 and 17 to properly support a rejection under 35 U.S.C. § 103. These failures of the cited art are accompanied by the Examiner's failure to acknowledge the clear advantages of the invention set forth in three declarations of unexpected results.

Soma and JP '913 have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition.



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Claim 16 is a method claim (corresponding to product claim 8 of Group VII) incorporating the limitations of claim 4 of Group I and claim 6 of Group IV, except that the material of the interconnector “comprises a material having a matrix of the general formula  $A_{1-x}B_xC_{1-y}D_yO_3$  where A is Mg, B is a rare earth element, aluminum or chromium, C is titanium, D is niobium or tantalum,  $0 < x \leq 0.2$  and  $0 \leq y \leq 0.2$ .” Claim 16 also contains the step of “integrally burning.”

In paragraph 4 of the Office Action mailed October 26, 2001, the Examiner admits the failure of the prior art of Soma to disclose the formula of claim 8 (and claim 6). The Examiner then asserts “Regarding the subscript ranges of the  $(La_{1-x}D_x)_{1-u}B_{1-w}O_3$  material, these ranges have not been shown to be critical variables in the practice of the invention. Applicant must show that the particular subscript ranges are critical, generally by showing that the claimed ranges achieve unexpected results relative to the prior art ranges.” (citation omitted).

However, Fig. 30 of the application shows the criticality of the  $x = 0.2$  limitation, and this result is directly opposite the teachings at column 4, lines 44 and 45 of Soma. A prior art reference must be considered in its entirety, i.e., as a whole, including portions that would lead away from the claimed invention. W.L. Gore & Associates, Inc. v. Garlock, Inc.



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Regarding integrally burning, in paragraph 3 of the Office Action mailed July 17, 2002 (Paper No. 24), the Examiner analogizes the heat treatment temperature of Soma to the inventive integrally burning step. However, the logical failures of this analogy have been discussed above. That is, if this heat treatment step of Soma is an integrally burning or sintering step, why use thermal spraying at all? That is, the Examiner is inferring that two discrete steps are needed in Soma to form the material. In contrast, the present invention only requires one step: sintering. Therefore, the Examiner is either 1) verifying that Soma uses a fundamentally different process from the invention, or 2) the principal of operation must be changed. See *In re Ratti*. In either case, the rejection must fall by the Examiner's own admissions.

Therefore, claims 16 and 17 of Group VVIII are patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejection based on the claimed combination of limitations and the above arguments is respectfully requested.

**S. Group XIX, Claim 20**

JP '913 and Soma fail to suggest all of the elements set forth in claim 20 to properly support the rejection of Group XIX under 35 U.S.C. § 103.

JP '913 and Soma have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid



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repetition. Also, claim 20 depends upon claim 16 (Group XVIII), and all of the distinctions of the invention over Group XVIII are equally applicable to Group XIX.

Claim 20 further recites the current passage of the interconnector is current collection in the vertical direction. This embodiment is supported at page 5, lines 13-15 of the specification.

In paragraph 5 of the Office Action mailed April 10, 2000 (Paper No. 8), the Examiner asserts that "the direction of current collection is a design choice that may be manipulated according to the needs of the artisan." The Examiner repeats this assertion in paragraph 4 of the Office Action mailed October 26, 2001 (paper No. 21).

However, as noted at page 8 of the Amendment filed July 10, 2002, vertical collection of current would not be an obvious design choice for one of ordinary skill in the art of producing fuel cells. Figures 44(a) and 44(b) of the instant invention depict vertical and horizontal current collection. As shown in Figure 44(a) and described on page 34, lines 8-10 of the specification, vertical current collection has advantages because high resistance can be evaded by the thinness of the interconnector. In vertical collection of current, the air electrode 15 is located just above the fuel electrode 12 via the interconnector 14. By thinning the layer of the interconnector, overvoltage due to resistance of the interconnector is decreased.





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In contrast, Figure 44(b) of the application shows that current collection in the horizontal direction is disadvantageous because it is not applicable for high resistance material. In horizontal current collection, current needs to be passed through the thin film in the same direction as the axial direction. Thus, a decrease in overvoltage cannot be achieved.

As discussed above, horizontal current collection and vertical current collection are different processes which possess different properties. Thus, the vertical direction of the fuel cells of the instant invention is not an arbitrary design choice.

Therefore, claim 20 of Group XIX is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejection based on the claimed combination of limitations and the above arguments is respectfully requested.

**T. Group XX, Claim 23**

JP '913 and Soma fail to suggest all of the elements set forth in claim 23 to properly support the rejection of Group XX under 35 U.S.C. § 103.

JP '913 and Soma have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition. Also, claim 22 depends upon claim 16 (Group XVIII), and all of the distinctions of the invention over Group XVIII are equally applicable to Group XX.



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Claim 23 further covers the embodiment that integrally burning is performed at a temperature of 1,300 °C to 1,400 °C. This advantageous temperature is discussed at page 6, lines 11-13 of the specification, which states: "This material can be burned not at a conventional high temperature of 1,600 °C, but at a lower temperature of 1,300 to 1,400 °C . . ."

In paragraph 3 of the Office Action mailed July 17, 2002 (Paper No. 24), the Examiner analogizes the heat treatment temperature of Soma to the inventive integrally burning step. However, the logical failures of this analogy have been discussed above. That is, if this heat treatment step of Soma is an integrally burning or sintering step, why use thermal spraying at all? That is, the Examiner is inferring that two discrete steps are needed in Soma to form the material. In contrast, the present invention only requires one step: sintering. Therefore, the Examiner is either 1) verifying that Soma uses a fundamentally different process from the invention, or 2) the principal of operation must be changed. See In re Ratti. In either case, the rejection must fall by the Examiner's own admissions.

Therefore, claim 23 of Group XX is patentable for these additional reasons as well. Accordingly, reversal of the Examiner's rejection based on the claimed combination of limitations and the above arguments is respectfully requested.



Appl. No.: 09/118,833

**II. Group XXI, Claim 28**

Soma or the combination of Soma and JP '913 fails to suggest all of the elements set forth in claim 28 to properly support the rejections of Group XXI under 35 U.S.C. § 103.

Soma and JP '913 have been discussed above, and the general discussion thereof is incorporated here, but is not being repeated here so as to avoid repetition.

Claim 28 of Group XX is similar to claim 4 of Group I with the exception the matrix is "consisting essentially of"  $\text{MtiO}_3$  where M is Mg, Ca, Sr, or Ba. That is, all the arguments and evidence for the patentability of claim 4 of Group I are equally applicable for claim 28 of Group XX. The arguments and evidence are hereby incorporated but not repeated for the sake of brevity.

Further, the "consisting essentially of" limitation renders this embodiment more restrictively defined over the prior art. As a result, the patentability of claim 28 of Group XX is entirely clear.

Accordingly, reversal of the Examiner's rejections based on the claimed combination of limitations and the above arguments and evidence is respectfully requested.

**V. Conclusion**

Appellants have demonstrated that the Examiner has failed to successfully allege that the rejected claims are *prima facie* obvious. Further, even if the Examiner established a *prima facie* case of obviousness, then this obviousness is rebutted by unexpected results.

For the reasons advanced above, it is respectfully submitted that all claims in this application are allowable. Thus, favorable reconsideration and reversal of the Examiner's rejection of claims 4-28 under 35 U.S.C. § 103, by the Honorable Board of Patent Appeals and Interferences, are respectfully solicited.

The required Appeal Brief fee in the amount of \$320.00 is attached hereto.

Pursuant to 37 C.F.R. §§ 1.17 and 1.136(a), Applicant(s) respectfully petition(s) for a two (2) month extension of time for filing a reply in connection with the present application, and the required fee of \$410.00 is attached hereto.

If necessary, the Commissioner is hereby authorized in this, concurrent, and further replies, to charge payment or credit any overpayment to Deposit

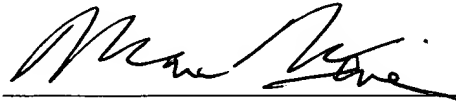
Appl. No.: 09/118,833

Account No. 02-2448 for any additional fee required under 37 C.F.R. § 1.16 or under 37 C.F.R. § 1.17; particularly, extension of time fees.

Respectfully submitted,

BIRCH, STEWART, KOLASCH & BIRCH, LLP

By:



Marc S. Weiner  
Reg. No.: 32,181

MSW/REG/jmb  
Attachment: APPENDIX A  
APPENDIX B  
APPENDIX C  
APPENDIX D

P.O. Box 747  
Falls Church, VA 22040-0747  
(703) 205-8000



APPENDIX A  
CLAIMS ON APPEAL

4. A solid electrolyte fuel battery, in which a sintered interconnector is used for connecting cells of the solid electrolyte fuel battery, and the sintered interconnector comprises a material having a matrix of the general formula  $\text{MTiO}_3$  where M is Mg, Ca, Sr, or Ba.

5. The solid electrolyte fuel battery as claimed in claim 4, wherein the current passage of the interconnector is current collection in the vertical direction from a fuel electrode through the interconnector.

6. A solid electrolyte fuel battery, in which a co-sintered interconnector for connecting cells of the solid electrolyte fuel battery comprises a material having a matrix of the general formula  $\text{A}_{1-x}\text{B}_x\text{C}_{1-y}\text{D}_y\text{O}_3$  where A is Ca, Sr or Ba, B is a rare earth element, aluminum or chromium, C is titanium, D is niobium or tantalum,  $0 < x \leq 0.2$  and  $0 \leq y \leq 0.2$ .

7. The solid electrolyte fuel battery as claimed in claim 6, wherein the current passage of the interconnector is current collection in the vertical direction.



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8. A solid electrolyte fuel battery, in which a co-sintered interconnector for connecting cells of the solid electrolyte fuel battery comprises a material having a matrix of the general formula  $A_{1-x}B_xC_{1-y}D_yO_3$  where A is Mg, B is a rare earth element, aluminum or chromium, C is titanium, D is niobium or tantalum,  $0 < x \leq 0.2$  and  $0 \leq y \leq 0.2$ .

9. The solid electrolyte fuel battery as claimed in claim 8, wherein the current passage of the interconnector is current collection in the vertical direction.

10. A solid electrolyte fuel battery, in which an interconnector for connecting cells of the solid electrolyte fuel battery comprises a material having a matrix of the general formula  $MTiO_3$  where M is Mg, Ca, Sr, or Ba, wherein the interconnector is integrally burned within said battery.

11. The solid electrolyte fuel battery as claimed in claim 10, wherein said battery comprises a fuel electrode, an electrolyte, an interconnector and an air electrode laminated onto a substrate, which are integrally burned within said battery.

12. A method of making a solid electrolyte fuel battery, in which an interconnector for connecting cells of the solid electrolyte fuel battery is co-sintered, and comprises a material having a matrix of the general formula



Appl. No.: 09/118,833

MTiO<sub>3</sub> where M is Mg, Ca, Sr, or Ba, said method comprising:

integrally burning within said battery the interconnector for connecting cells of the solid electrolyte fuel battery.

13. The method of making the solid electrolyte fuel battery as claimed in claim 12, wherein said battery comprises a fuel electrode, an electrolyte, an interconnector and an air electrode laminated onto a substrate.

14. A method of making a solid electrolyte fuel battery, in which a co-sintered interconnector for connecting cells of the solid electrolyte fuel battery comprises a material having a matrix of the general formula  $A_{1-x}B_xC_{1-y}D_yO_3$  where A is Ca, Sr or Ba, B is a rare earth element, aluminum or chromium, C is titanium, D is niobium or tantalum,  $0 < x \leq 0.2$  and  $0 \leq y \leq 0.2$ , said method comprising:

integrally burning within said battery the interconnector for connecting cells of the solid electrolyte fuel battery.

15. The method of making the solid electrolyte fuel battery as claimed in claim 14, wherein said battery comprises a fuel electrode, an electrolyte, an interconnector and an air electrode laminated onto a substrate.





16. A method of making a solid electrolyte fuel battery, in which a co-sintered interconnector for connecting cells of the solid electrolyte fuel battery comprises a material having a matrix of the general formula  $A_{1-x}B_xC_{1-y}D_yO_3$  where A is Mg, B is a rare earth element, aluminum or chromium, C is titanium, D is niobium or tantalum,  $0 < x \leq 0.2$  and  $0 \leq y \leq 0.2$ , said method comprising:

integrally burning within said battery the interconnector for connecting cells of the solid electrolyte fuel battery.

17. The method of making the solid electrolyte fuel battery as claimed in claim 16, wherein said battery comprises a fuel electrode, an electrolyte, an interconnector and an air electrode laminated onto a substrate.

18. The method of claim 12, wherein the current passage of the interconnector is current collection in the vertical direction.

19. The method of claim 14, wherein the current passage of the interconnector is current collection in the vertical direction.

20. The method of claim 16, wherein the current passage of the interconnector is current collection in the vertical direction.



Appl. No.: 09/118,833

21. The method of claim 12, wherein the integrally burning is performed at a temperature of 1,300 °C to 1,400 °C.

22. The method of claim 14, wherein the integrally burning is performed at a temperature of 1,300 °C to 1,400 °C.

23. The method of claim 16, wherein the integrally burning is performed at a temperature of 1,300 °C to 1,400 °C.

24. The solid electrolyte fuel battery as claimed in claim 4, wherein the interconnector is a hermetic interconnector having a relative density of greater or equal to 94%.

25. The solid electrolyte fuel battery as claimed in claim 6, wherein the interconnector is a hermetic interconnector having a relative density of greater or equal to 94%.

26. The solid electrolyte fuel battery as claimed in claim 8, wherein the interconnector is a hermetic interconnector having a relative density of greater or equal to 94%.

27. The solid electrolyte fuel battery as claimed in claim 10, wherein the interconnector is a hermetic interconnector having a relative density of greater or equal to 94%.



Appl. No.: 09/118,833

28. A solid electrolyte fuel battery, in which a sintered interconnector is used for connecting cells of the solid electrolyte fuel battery, and the sintered interconnector comprises a material having a matrix consisting essentially of  $\text{MTiO}_3$  where M is Mg, Ca, Sr, or Ba.



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of :  
Toshiro NISHI, et al. : Group Art Unit: 1745  
Serial Number: 09/118,833 : Examiner: CREPEAU. J  
Filed: 07.20.1998 :  
For: SOLID ELECTROLYTE TYPE FUEL BATTERY

**Declaration under 37 CFR 1.132**

I. I, Toshiro NISHI, declare

1. That I am the same person who executed the Declaration filed in the patent of the above-identified application;  
that I have been employed by MITSUBISHI HEAVY INDUSTRIES, Ltd. for 18 years and 10 months, and have worked at the Research Laboratory of the Company.
2. That I have conducted experiments in order to demonstrate unexpected effects of the invention described in United States Patent Application Serial No. 118,833 over compositions described in the cite U.S. Patent issued to SOMA et al.
3. That I have performed or supervised the performance of the experiments described below and have reviewed the results thereof as reported below.
4. That the conclusions stated herein are based upon this review of the results and my experience in this

field.

5. Toshiro NISHI, the declarant, has been involved in research into fuel batteries in the Research Laboratory of the Company for 17 years. He and his colleagues, Nobuaki MURAKAMI and Hirokazu YAMAMOTO, found a process to reduce the cost for producing fuel batteries and to simplify the production process by using an interconnector comprised of a specific composition, and applied for U.S. Patent in July 20, 1998.

In order to substantiate the difference from the cited patent issued to Soma et al., we hereby submit with tests results described below.

#### 6. Tests

(1) Fuel Battery prepared by Sintering Process (Present invention)

As the material,  $\text{MTiO}_3$ , which is concretely  $\text{Ca}_{0.9}\text{La}_{0.1}\text{TiO}_3$  as described in the claim 4 of the present invention, is used.

The detail procedure is accorded with the procedure described in Appendix A-1 attached herewith, "Evaluation of Interconnector Preparation Method of Interconnector", and Appendix A-2 attached herewith, "Performance of Sintered Cells and Module Manufacturing Process of Sintered Cell".

These references are parts of papers prepared on June, 1998 by the declarant for their lecture regarding fuel batteries prepared by employing Sintering process.

Namely, as written in the Appendix A-1,  $\text{CaCO}_3$ ,  $\text{TiO}_2$  and  $\text{La}_2\text{O}_3$  are incorporated and mixed by using a ball mill, and the mixture was then subjected to firing.

Then, as shown in the Appendix A-2, the slurry is printed in band pattern by means of screen printing method around the surface of the substrate 11 being prepared by extrusion molding.

At this stage, as shown in Fig. 44 in the description in the present patent application, a material for a fuel electrode 12 is laminated onto the side surface of the base tube 11, and a material for an electrolyte 13 and a material for an interconnector 14 are then laminated onto the fuel electrode in series, and the laminated materials were subjected to the first sintering process at approximately  $1,300^\circ\text{C}$ . Then, the slurry for an air electrode is printed in band pattern and is then subjected to the second sintering process at  $1,250^\circ\text{C}$  to obtain the cell.

## (2) Fuel Battery prepared by Thermal Spraying Process

In this process,  $\text{LaCrO}_3$  is used as the material.

More specifically, hybrid plasma spraying torch, which is described in Fig. 2 in the Appendix B, is used and the condition for the thermal spraying is accorded with that shown in the Table 2.

The temperature employed for the thermal spraying was  $2000^\circ\text{C}$ .

When employing the thermal spraying process, it requires very laborious steps, i.e., thermally spraying a material

for a fuel electrode onto a substrate 11 to form a fuel electrode 12, then applying masking and thermally spraying a material for an electrolyte to form an electrolyte 13, then applying masking and thermally spraying a material for an interconnector to form an interconnector 14, and finally applying masking and thermally spraying a material for an air electrode to obtain an air electrode 15.

(3) The following is a comparative difference in between the fuel battery prepared by sintering process and thermal spraying process.

	Fuel Cell prepared by Thermal Spraying Process	Fuel Cell prepared by Sintering Process (Present Invention)
Required time for Production	150 min/fuel cell	15 min/fuel cell
Yield on materials	3 - 10%	90% or more
Equipment cost	Basic amount	1/10
Construction cost	Basic amount	1/5
Materials cost	Basic amount	1/2
Cell production cost	1.3 million yen/kw	50,000 yen/kw

As shown above, the sintering process of the present invention can markedly decrease the production cost in comparison with the thermal spraying process of the citation. This is neither disclosed nor suggested in the citation SOMA et al .

In particular, in case of the thermal spraying process, the yield on materials is very low since most of the materials are consumed as useless during the thermal spraying. While in case of the sintering process according to the present invention, the yield on materials is improved as much as

10-30 times comparing to the thermal spraying process, which contributes to greatly decrease the materials cost.

Furthermore, in the thermal spraying process described above, no heat treatment at 1,250°C or higher is applied following to the thermal spraying as done in the citation of Soma et al., and therefore, the equipment cost for applying heat treatment must be further required and more time will be required for the production in case of employing the thermal spraying process.

In addition, according to the present invention, an advantageous effect of improved impaction property can be attained based on strong bonding force given by the chemical co-sinter in which oxygen (O) is shared.

On the other hand, in case of the thermal spraying process, the materials tend to be easily exfoliated because the bonding is provided in a form of weak intermolecular force such as static electricity.

This advantageous characteristic provided by the present invention is an essential feature to be used as a fuel cell which is required to be used over a long time.

Accordingly, the material for the interconnector in a fuel cell prepared by the thermal spraying process, which is remarkably different from the production process in the present invention, cannot necessarily be directly used for the interconnector in a fuel cell prepared by the sintering process. That is, the fact that  $\text{MtTiO}_3$ , which is expressed



as  $\text{Ca}_{0.9}\text{La}_{0.1}\text{TiO}_3$  in the present declaration letter, is suitable for the material to be used for an interconnector in a sinter type fuel cell was found for the first time by the inventors of the present invention.

II. The undersigned further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and, further, that these statements were made with knowledge that willful, false statements, and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of title 18 of the United State Code and that such willful, false statements may jeopardize the validity of the above-identified application or any patent issuing thereon.

Toshiro Nishi

Toshiro Nishi

Dated: This 2<sup>nd</sup> day of February, 2001

Appendix:

Appendix A-1

Appendix A-2

Appendix B

# EVALUATION of Interconnector Preparation Method of Interconnector

MO or  $\text{MCO}_3$

( $\text{CaCO}_3$  etc)

$\text{TiO}_2$

$\text{Ln}_2\text{O}_3$

( $\text{La}_2\text{O}_3$  etc)

Mixing (Ball Mill, 24h)

Calcination (1473K X 5h)

Grinding (Ball Mill, 48h)

Forming (CIP, 200MPa)

Sintering



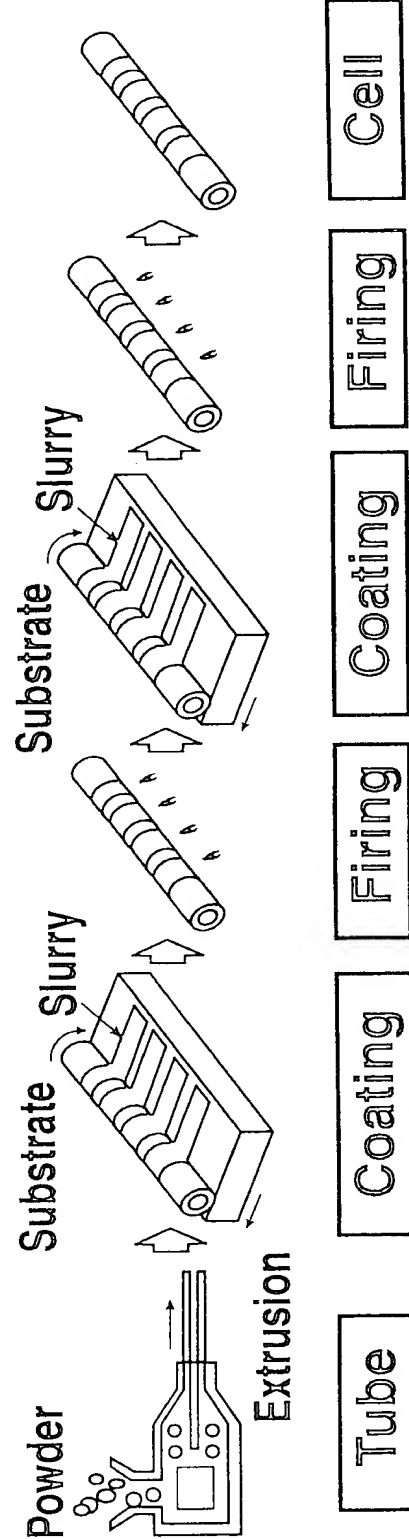
Manufacturing of  
Sintered Cell  
(Next Page)

Characterization



mitsubishi  
HEAVY INDUSTRIES, LTD.

# PERFORMANCE of Sintered Cells and Module Manufacturing Process of Sintered Cell



## POINT

- ① High Yield → Reduction of Material Cost
- ② Non Masking → Reduction of Process
- ③ Simple Facilities → Reduction of Equipment Investment

## EFFECT

STUDY ON R.F. PLASMA SPRAYING

- A Newly Developed Process of Plasma Spraying -

Akira Notomi\*, Yasuyuki Takeda\*, Masaru Kodama\* and Masayoshi Shitamichi\*

ABSTRACT

A new plasma spraying process has been developed, which utilizes a radio frequency induced plasma or a hybrid plasma of r.f. and d.c. arc plasma. The r.f. plasma has a wider controlling range of plasma than that of the conventional d.c. arc plasma. The r.f. plasma and hybrid plasma spraying of  $ZrO_2$  and  $LaCrO_3$  particles were studied, stabilizing the plasma condition. It is indicated that this new plasma spraying process has a potential to produce a dense coating of ceramics.

1. INTRODUCTION

Thermal spraying has been applied in a wide range for wear, corrosion and heat resistant coatings. Especially in thermal spraying processes, d.c. arc plasma (d.c. plasma) spraying is the most promising one, which is able to produce the coatings of refractory metals and ceramics because of its high temperature plasma. However, the high temperature region of d.c. plasma is so narrow and gas velocity is so high that there would be many unmelted particles in the coating. This could cause to decrease the adhesive, cohesive strength and density of coating.

On the other hand, radio frequency plasma (r.f. plasma) has wide high temperature region of several cm diameter, and low gas velocity, which could melt the spraying particles sufficiently<sup>1)</sup>. Hybrid plasma of r.f. and d.c. plasma could widen a plasma controlling range and is considered to have potential to produce a denser coating of new material which have been difficult to produce by conventional process<sup>2)</sup>.

In this study, r.f. plasma torch and hybrid plasma torch were trially manufactured and the stabilizing conditions of plasma generation were obtained. Using these plasma torches, plasma spraying of ceramics was carried out, by which result it is confirmed that these new plasma processes could produce a denser and functional coatings.

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\* Nagasaki R & D Center, Mitsubishi Heavy Industries, Ltd. Japan

## 2. EXPERIMENTAL PROCEDURE

Fig. 1 shows a trially manufactured r.f. plasma torch. A water cooled quartz tube of inner diameter 57 mm is set in r.f. coil of 3 turns. Sheath gas and carrier gas flow into the quartz tube from gas nozzle which located in upper side of quartz tube. Sheath gas has two flows of radial and tangential direction which effect on stabilizing a plasma and cooling a quartz tube. Particles can be supplied into quartz tube throughout center of gas nozzle by carrier gas.

Fig. 2 shows a trially manufactured hybrid plasma torch where d.c. plasma torch is set on the upper side of quartz tube in place of gas nozzle used in r.f. plasma torch. Sheath gas of radial direction flows into the quartz tube. Particles are supplied to the tip of d.c. plasma torch.

Input power and frequency of r.f. generator used are 130 KVA max., 4 MHz respectively. Commercial d.c. plasma torch of input power 30 KVA, nozzle diameter 5 mm is used for hybrid plasma torch. It is important to know the operating conditions to stabilize a plasma because of absence of electrode in r.f. plasma torch. The appropriate conditions of gas flow rate and input power are studied concerning with both of r.f. and hybrid plasma.  $\text{ZrO}_2\text{-12wt\%Y}_2\text{O}_3$  (YSZ) particles (30~50  $\mu\text{m}$ ) were r.f. plasma sprayed on the substrate varying the distance from torch. The diameters of flattened particles were measured.  $\text{LaCrO}_3$  particles (20~85  $\mu\text{m}$ ) were also r.f. and hybrid plasma sprayed on the substrate, and the coatings were compared with d.c. plasma sprayed one.

## 3. RESULTS AND DISCUSSION

Fig. 3 shows the r.f. plasma operating conditions to maintain a stabilized plasma concerning with the relation between r.f. input power and  $\text{H}_2$  flow rate as secondary gas which is supplied to sheath gas of radial direction. Fig. 4 also shows the hybrid plasma operating condition concerning with the relation between r.f. input power and d.c. input power.

Based on Fig. 3, r.f. plasma spraying conditions were determined as shown in Table 1. Fig. 5 shows the relation between flattened particle diameter and ratio of particle number as parameter of distance from torch to substrate. There is maximum ratio of particle number at the diameter of 70~90  $\mu\text{m}$  (flattened ratio: 1.5~2.0) independently of the distance from torch to substrate. The maximum value of the ratio of particle number is obtained at  $d=45$  mm. Fig. 6 shows SEM photographs of the surface of YSZ coating by the above conditions. There can be seen widely flattened particles in r.f. plasma sprayed coating, while the flattened ratio is not so high in the conventional d.c. plasma sprayed coating. However, the adhesive strength of particles in r.f. plasma sprayed coating is lower than that of

the latter one, which is considered that the kinetic energy of particles in r.f. plasma is not sufficient for dense coating of ceramics.

Hybrid plasma would have a potential to be an effective process to solve this problem. Hybrid plasma spraying of  $\text{LaCrO}_3$  was carried out using the spraying conditions as shown in Table 2. The r.f. and d.c. plasma spraying were also carried out to compare with that. Fig. 7 shows SEM photographs of surfaces and fracture surfaces of the coatings by three processes. Hybrid plasma sprayed coating shows the highest flattened ratio of particle and seems to be the densest one from fracture surface. There also can be seen flattened particles in r.f. plasma coating, of which flattened ratio is not so high. The flattened particles and dispersed fine particles are observed on the surface of d.c. plasma sprayed coating, of which fracture surface does not show a dense structure. These results show that hybrid plasma spraying could produce a denser ceramic coating than the other plasma spraying process.

It should be necessary to take into account of characteristics of spraying material and coating function needed, in applying r.f. plasma or hybrid plasma spraying process effectively.

#### 4. CONCLUSIONS

The r.f. plasma and hybrid plasma spraying of ceramics which have wide controlling range of plasma were carried out as new plasma spraying processes. The results obtained were summarized as follows:

- (1) The r.f. plasma sprayed particles of  $\text{ZrO}_2\text{-12wt\%Y}_2\text{O}_3$  were sufficiently flattened on the surface of substrate, which would show possibility to produce a dense ceramic coating. However, the adhesive strength of particles was not so good under the spraying conditions used.
- (2) Hybrid plasma spraying of  $\text{LaCrO}_3$  produces denser coating than r.f. and d.c. plasma spraying process, which would show that hybrid plasma spraying process has potential to produce dense coating of high melting point material and ceramics.

#### REFERENCES

- 1) S. Takeuchi, T. Okada, T. Yoshida and K. Akashi: J. Japan Inst. Metals, 52, 711 (1988).
- 2) T. Okada, H. Hamatani and T. Yoshida: J. Am. Ceram. Soc. 72[11] 2111-16 (1989).

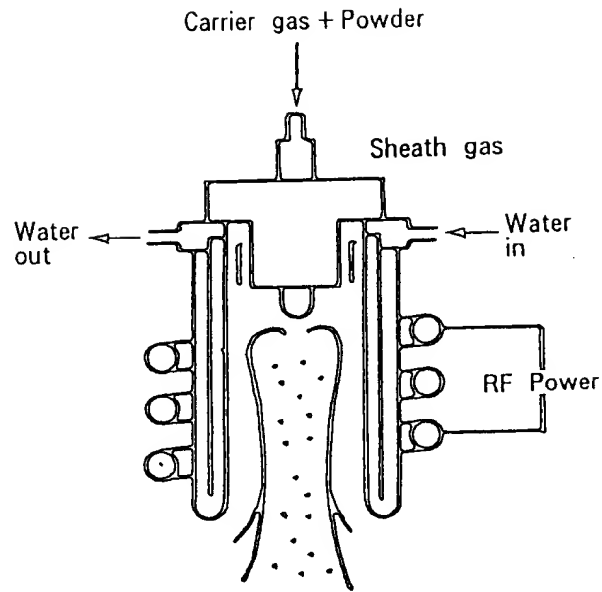


Fig.1 R.F. plasma spraying torch

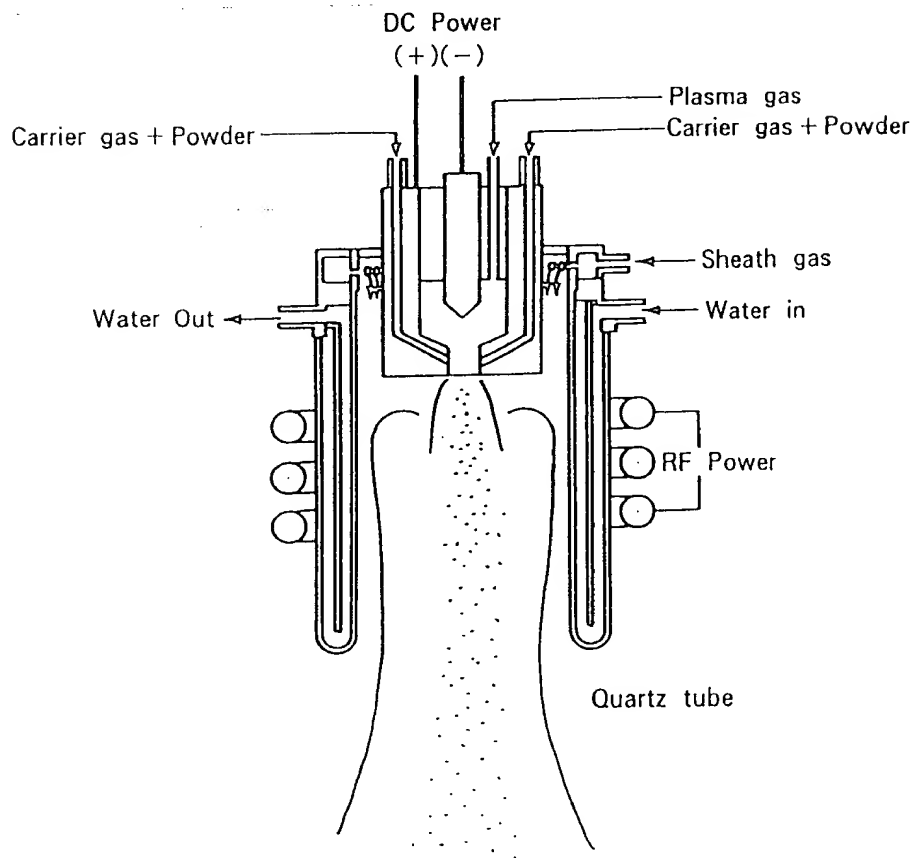


Fig.2 Hybrid plasma spraying torch

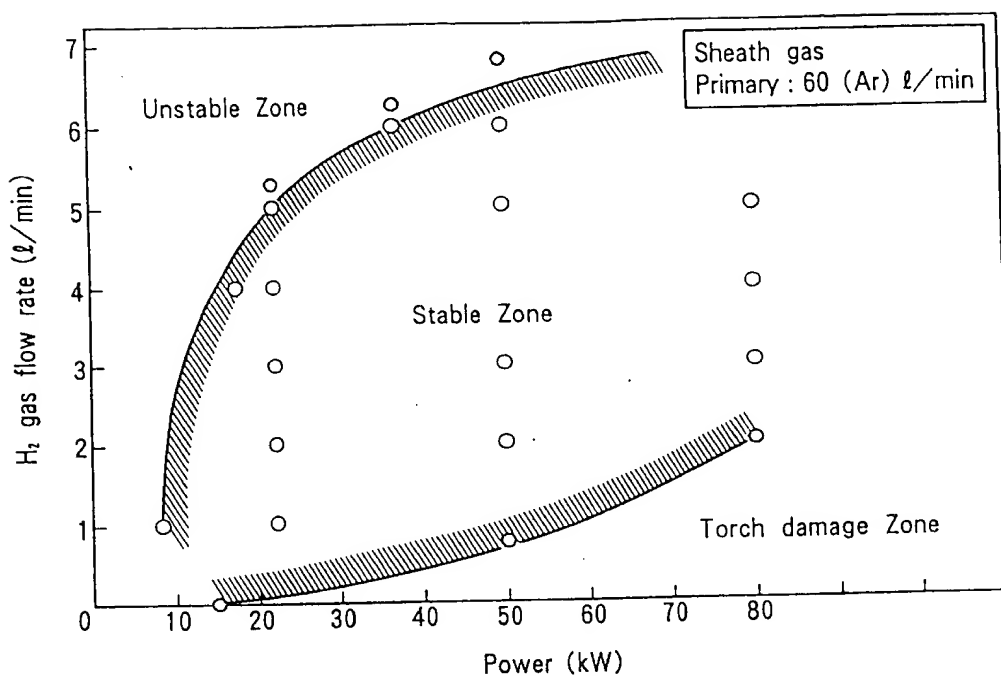


Fig.3 Effect of secondary gas ( $H_2$ ) flow rate on stability of R.F. plasma

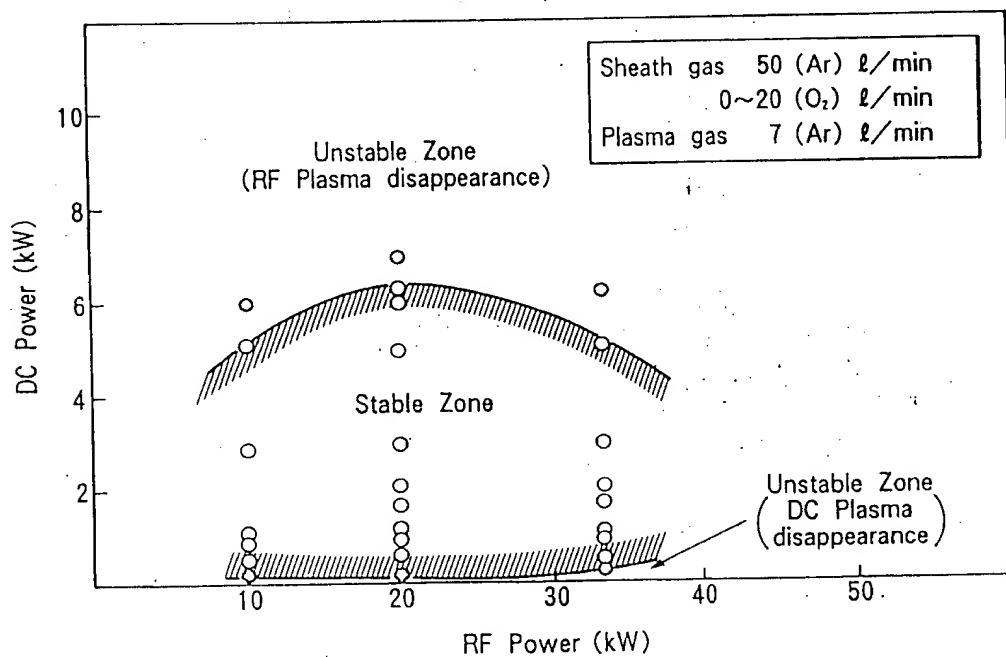


Fig.4 Effect of R.F. and D.C. power on stability of hybrid plasma



Table 1 Conditions of R.F. plasma spraying

Material	Size ( $\mu\text{m}$ )	RF Plate Power (kW)	Sheath gas ( $\ell/\text{min}$ )	Carrier gas ( $\ell/\text{min}$ )	Powder feed ( $\text{g}/\text{min}$ )	Spraying Distance (mm)
$\text{ZrO}_2$ -15(wt) $\text{Y}_2\text{O}_3$	30~50	50	50 (Ar) 5 ( $\text{H}_2$ )	9 (Ar)	10	5~45

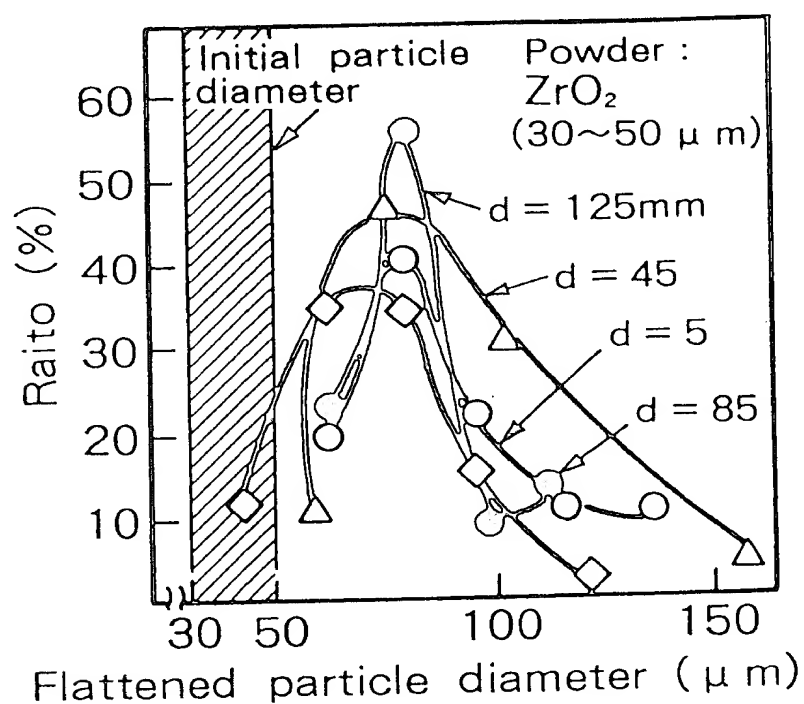
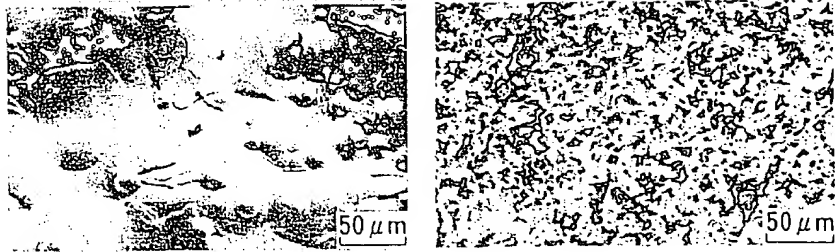


Fig.5 Distribution of flattened ratio of R.F. plasma sprayed particles



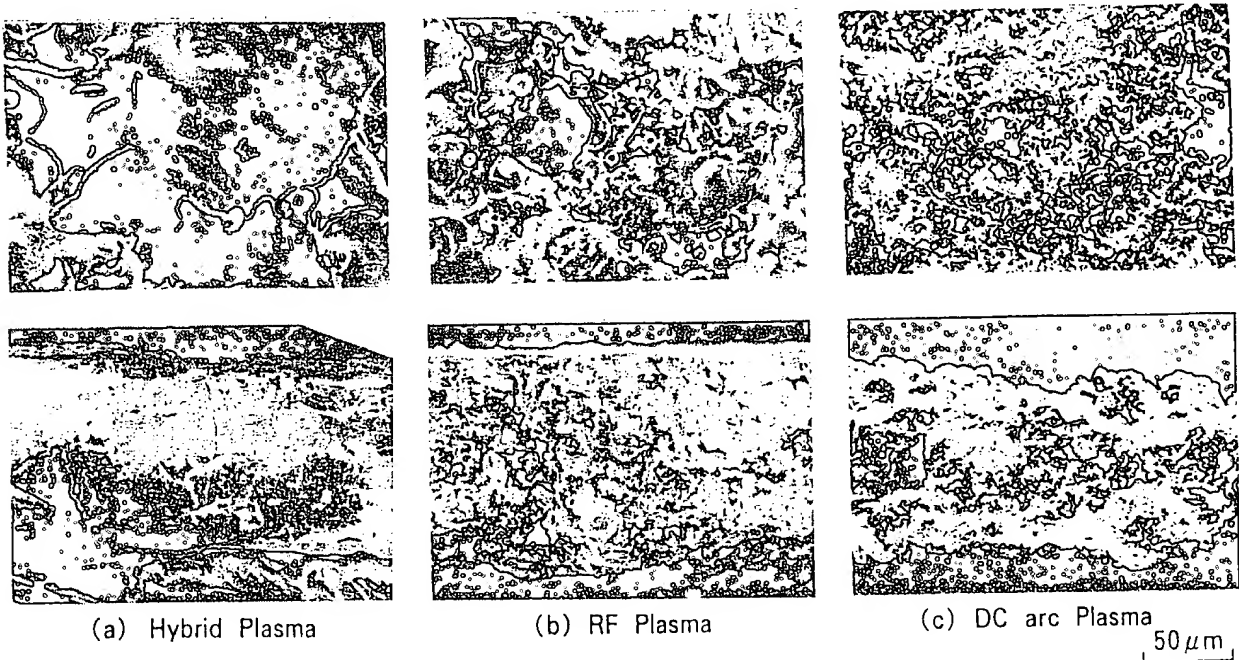
(a) RF Plasma

(b) DC arc Plasma

Fig.6 Surface microstructures of R.F. and D.C. plasma sprayed coatings of YSZ

Table 2 Conditions of hybrid plasma spraying

Material	Size ( $\mu\text{m}$ )	Power (kW)		Gas flow rate ( $\ell/\text{min}$ )			Powder feed ( $\text{g}/\text{min}$ )	Spraying distance (mm)
		RF Plate	DC	Sheath gas	Plasma gas	Carrier gas		
$\text{LaCrO}_3$	20~85	20	7	50 (Ar)	5 (Ar)	3.5 (Ar)	1	10



(a) Hybrid Plasma

(b) RF Plasma

(c) DC arc Plasma

50  $\mu\text{m}$

Fig.7 Surface and fracture surface microstructures of plasma sprayed coatings of  $\text{LaCrO}_3$



APPENDIX C

PATENT  
0965-0232P

IN THE U.S. PATENT AND TRADEMARK OFFICE

Applicants: Toshio NISHI et al. Conf.: 9403  
Serial No.: 09/118,833 Art Unit: 1745  
Filed: July 20, 1998 Examiner: J. CREPEAU  
For: SOLID ELECTROLYTE TYPE FUEL BATTERY

**DECLARATION UNDER 37 C.F.R. §1.132**

Assistant Commissioner for Patents  
Washington, DC 20231

Sir:

I, Toshio NISHI, do declare and say as follows:

1. I am one of the inventors of the U.S. Patent Application Serial No. 09/118,833, filed July 20, 1998, and I am familiar with the contents of the application, its prosecution before the U.S. Patent and Trademark Office, and the references cited therein. I am a citizen of Japan, residing at c/o Nagasaki Research & Development Center, Mitsubishi Heavy Industries, Ltd., 717-1, Fukahori-machi 5-chome, Nagasaki-shi, Nagasaki, Japan. I have been employed by Mitsubishi Heavy Industries, Ltd. for over 20 years, and have worked at the Research Laboratory of the Company.

2. I have studied the contents of U.S. Patent 5,411,767, hereinafter referred to as SOMA.

3. To show the superiority of the present invention, I am submitting the attached SEM photomicrographs of plasma sprayed material (A) and co-sintered material (B). In reference to these SEM photomicrographs (A) and (B), I am making the following observations:

When plasma spraying is applied as in SOMA and other conventional art processes, a number of air spaces are produced in the plasma-sprayed film having a thickness of from several to dozens of microns. This can be clearly observed in the dense-blue cored portion of SEM photomicrograph (A) (also note the micron bar for scale).

In contrast, when the inventive co-sintering is utilized, a film having a very dense structure is formed. This is clearly observable in SEM photomicrograph (B). Incidentally, the portions that appear to be air spaces in SEM photomicrograph (B) are produced during thermal etching. However, the sizes of these air spaces are very small, on the order of submicrons, as indicated by the micron bar for scale.

As a result, a "sintered" or "co-sintered" material such as an interconnector does not represent a process step. Instead, a "sintered" or "co-sintered" material represents a physical state of matter that is achieved through the sintering process. This state of matter is readily discernable by comparing SEM photomicrographs (A) and (B).

Therefore, the plasma sprayed film of SOMA is incapable of attaining the sintered or co-sintered interconnector of the invention. Therefore, SOMA fails to either teach or suggest a sintered or co-sintered interconnector of the invention. The high density achievable by the inventive technology and demonstrated in the attached SEM photomicrographs are a clear demonstration of unexpected results over SOMA.

4. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

October 29, 2002  
Date

By Toshiro Nishi  
Toshiro NISHI



PATENT  
0965-0232P

IN THE U.S. PATENT AND TRADEMARK OFFICE

Applicants: Toshio NISHI et al. Conf.: 9403  
Serial No.: 09/118,833 Art Unit: 1745  
Filed: July 20, 1998 Examiner: J. CREPEAU  
For: SOLID ELECTROLYTE TYPE FUEL BATTERY

DECLARATION UNDER 37 C.F.R. §1.132

Assistant Commissioner for Patents  
Washington, DC 20231

Sir:

I, Toshio NISHI, do declare and say as follows:

1. I am one of the inventors of the U.S. Patent Application Serial No. 09/118,833, filed July 20, 1998, and I am familiar with the contents of the application, its prosecution before the U.S. Patent and Trademark Office, and the references cited therein. I am a citizen of Japan, residing at c/o Nagasaki Research & Development Center, Mitsubishi Heavy Industries, Ltd., 717-1, Fukahori-machi 5-chome, Nagasaki-shi, Nagasaki, Japan. I have been employed by Mitsubishi Heavy Industries, Ltd. for over 20 years, and have worked at the Research Laboratory of the Company.

2. I have studied the contents of U.S. Patent 5,411,767, hereinafter referred to as SOMA.

3. To show the superiority of the present invention, I am submitting the attached SEM photomicrographs of plasma sprayed material (A) and co-sintered material (B). The materials (A) and (B) being compared have the same chemical composition. In reference to these SEM photomicrographs (A) and (B), I am making the following observations:

When plasma spraying is applied as in SOMA and other conventional art processes, a number of air spaces are produced in the plasma-sprayed film having a thickness of from several to dozens of microns. This can be clearly observed in the dense-blue cored portion of SEM photomicrograph (A) (also note the micron bar for scale).

In contrast, when the inventive co-sintering is utilized, a film having a very dense structure is formed. This is clearly observable in SEM photomicrograph (B). Incidentally, the portions that appear to be air spaces in SEM photomicrograph (B) are produced during thermal etching. However, the sizes of these air spaces are very small, on the order of submicrons, as indicated by the micron bar for scale.

As a result, a "sintered" or "co-sintered" material such as an interconnector does not represent a process step. Instead, a "sintered" or "co-sintered" material represents a physical state of matter that is achieved through the sintering process. This state

of matter is readily discernable by comparing SEM photomicrographs (A) and (B).

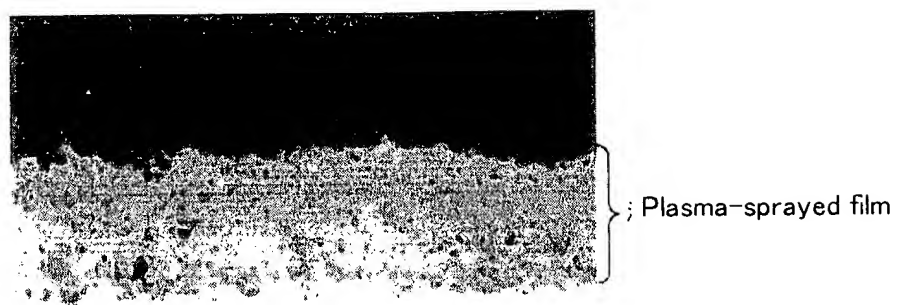
Therefore, the plasma sprayed film of SOMA is incapable of attaining the sintered or co-sintered interconnector of the invention. Therefore, SOMA fails to either teach or suggest a sintered or co-sintered interconnector of the invention. The high density achievable by the inventive technology and demonstrated in the attached SEM photomicrographs are a clear demonstration of unexpected results over SOMA.

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January 27, 2003  
Date

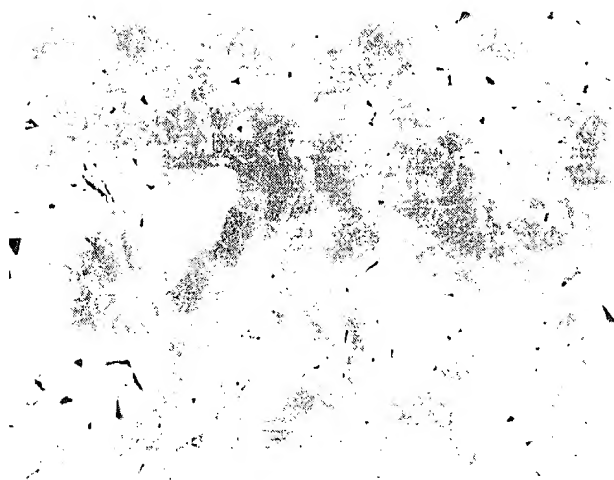
By Toshiro Nishi  
Toshiro NISHI





100  $\mu\text{m}$

(A)



(B)